



Europäisches Patentamt
European Patent Office
Office européen des brevets

(19)

(11) Publication number:

0 120 562
A2

(12)

EUROPEAN PATENT APPLICATION

(21) Application number: 84300497.9

(51) Int. Cl.³: C 08 L 23/16

(22) Date of filing: 26.01.84

C 08 L 23/10

//(C08L23/10, 23/16), (C08L23/16,
23/10)

(30) Priority: 27.01.83 US 461510

(71) Applicant: Exxon Research and Engineering Company

P.O.Box 390 180 Park Avenue
Florham Park New Jersey 07932(US)

(43) Date of publication of application:
03.10.84 Bulletin 84/40

(72) Inventor: Hazelton, Donald Ross
89 Glenmere Drive
Chatham New Jersey(US)

(84) Designated Contracting States:
BE DE FR GB IT

(74) Representative: Dew, Melvyn John et al,
Esso Chemical Ltd. Esso Chemical Research Centre P.O.
Box 1
Abingdon Oxfordshire, OX13 6BB(GB)

(54) Elastomer, polypropylene blends for optically clear products.

(57) Composition blends consisting essentially of an ethylene
propylene elastomer and polypropylene, particularly a ran-
dom polypropylene reactor copolymer, are very effective for
forming optically clear products, particularly films.

EP 0 120 562 A2

1 ELASTOMER, POLYPROPYLENE BLENDS FOR OPTICALLY CLEAR
 PRODUCTS2 FIELD OF THE INVENTION

3 This invention relates to blend compositions for
4 optically clear products consisting essentially of an
5 ethylene propylene elastomer and polypropylene, preferably
6 a random reactor copolymer.

7 PRIOR ART

8 Optically clear film products find particular
9 utility in medical facilities, particularly where the clar-
10 ity is important for monitoring fluid intake and detecting
11 fluid contamination. Plastic vinyl materials have tra-
12 ditionally been utilized, but problems have been encount-
13 ered because of serious adverse reactions from the plasti-
14 cizers required because of their extractability and sus-
15 pected carcinogenicity.

16 The prior art has suggested numerous polyolefin
17 blends, e.g., U.S. Patent 3,515,775 and 4,087,485 to give
18 the desired film products. In general, these blends have
19 been unable to replace the vinyl plastics because of dif-
20 ficult reproducibility, poor optical quality, or the need
21 for calendering to provide clarity. A significant aspect
22 of the clarity requirement is that that property survive
23 autoclave sterilization.

24 SUMMARY OF THE INVENTION

25 It has now been found that blends consisting es-
26 sentially of an ethylene propylene elastomer, and polypro-
27 pylene, preferably a random polypropylene reactor copoly-
28 mer, provide films and other products which are both opti-
29 cally clear and flexible when rapidly quenched, thus over-
30 coming the difficulties of the prior art in replacing the
31 vinyl materials. It is surprising that these properties
32 are obtained, since plastic blends without plasticizers re-
33 quire high elastomer content to achieve desired flexibility
34 and introduction of an elastomer into a plastic blend pro-
35 duces poorer optical properties.

1 DETAILED DESCRIPTION OF THE INVENTION

2 The elastomer is a non-crosslinked ethylene pro-
3 pylene elastomer having a Mooney viscosity, ML (1 + 8) @
4 127°C in the range of about 10 to 40, preferably 15 to 25,
5 and at least 50 weight percent ethylene, up to 85 percent,
6 preferably about 60 to 80 percent, with the balance pre-
7 dominantly propylene. The term ethylene propylene elasto-
8 mer is intended to generically connote ethylene propylene
9 copolymers (EPM) and ethylene propylene terpolymers (EPDM).
10 Typical of the third monomers is a C₅-C₁₄ nonconjugated
11 diolefin. Non-limited examples of such nonconjugated di-
12 olefins include the following:

13 A. Straight chain acyclic dienes such as: 1,4-
14 hexadiene and 1,6-octadiene.

15 B. Branched acyclic dienes such as 5-methyl 1,4-
16 hexadienes, 3,7-dimethyl 1,6-octadiene, 3,7-dimethyl 1,7-
17 octadiene and mixed isomers of dihydromyrcene, and di-
18 hydrocimene.

19 C. Single ring alicyclic dienes such as 1,4-cyclo-
20 hexadiene, 1,5-cyclooctadiene and 1,5-cyclododecadiene.

21 D. Multi-ring alicyclic fused and bridged ring
22 diene such as: tetrahydroindene, methyl tetrahydroindene,
23 alkenyl, alkylidene, cycloalkenyl and cycloalkylidene
24 norbornenes such as 5-methylene-2-norbornene (MNB), 5-
25 ethylidene-2-norbornene (ENB), 5-propenyl-2-norbornene,
26 5-isopropylidene-2-norbornene, 5-(4-cyclopentenyl)-2-
27 norbornene, and 5-cyclohexylidene-2-norbornene.

28 Preferred is 5-ethylidene-2-norbornene (ENB).

29 These ethylene/higher alpha olefin copolymers
30 and terpolymers may be prepared by any conventional manner
31 and the preparation of same does not constitute part of the
32 instant invention.

33 Blends useful in this invention employ polypro-
34 pylene which can be a highly crystalline isotactic or
35 syndiotactic polypropylene. Also, the polypropylene can
36 be a copolymer, referred to as a random polypropylene

1 reactor copolymer, containing minor amounts of an alpha-
2 olefin comonomer of 2 to 16 carbon atoms. The level of
3 comonomer which can be utilized is about 1 to about 20
4 weight percent, preferably about 2 to about 18, most pref-
5 erably about 2 to about 15; a preferred comonomer is ethyl-
6 ene. These are art-recognized materials, see, e.g., "Modern
7 Plastics Encyclopedia 1981-1982," pages 532, 533. The ran-
8 dom reactor copolymer is particularly preferred because it
9 gives lower stiffness and better optical properties. Espe-
10 cially useful is the random copolymer containing about 3
11 percent ethylene and having a melt flow rate of 4.0 at
12 230°C (ASTM D1238 condition L).

13 The elastomer is employed in the blend in an
14 amount of about 1 to 80 weight percent, and the polypropyl-
15 ene in an amount of about 20 to 99 weight percent.

16 In addition to the active components, various in-
17 gredients can be incorporated into the composition in order
18 to achieve various cost and/or performance objectives in
19 specific end-use applications. For example, one can use
20 such materials as process aids (e.g., stearic acid), lubri-
21 cants (e.g., oleamide, oil), antiblocking aids, antioxidants,
22 foaming agents and non-extractible low MW polyethylene or
23 oxidized polyethylene waxes as slip agents. Polypropylene
24 and reactor copolymer can be blended as one phase to opti-
25 mize cost or performance properties (e.g., flexibility,
26 softening temperature, etc.).

27 Preparation of compositions of this invention,
28 i.e., the blend of polyolefin plastics and elastomer as de-
29 scribed above, can be achieved in several different ways.
30 The polyolefin plastics and elastomer are brought into in-
31 timate contact by, for example, dry blending these materi-
32 als and then passing the overall composition through a com-
33 pounder extruder. Alternatively, the polyolefin plastics
34 and elastomer can be fed directly to a mixing device such
35 as a compounding extruder, high shear continuous mixer,
36 two roll mill, an internal mixer such as a Banbury, etc.
37 The optional ingredients previously described can be added.

1 to the composition during this mixing operation. It is
2 also possible to achieve melt mixing in an extruder sec-
3 tion of a film line or in an injection or extrusion blow
4 molding machine. Overall, the objective is to obtain a
5 uniform dispersion of all ingredients and this is readily
6 achieved by inducing sufficient shear and heat to cause
7 the plastics component(s) to melt. However, time and tem-
8 perature of mixing should be controlled as is normally
9 done by one skilled in the art so as to avoid molecular
10 weight degradation or volatilization of any of the ingredients.

11 The blends of the invention can be used to pro-
12 duce films, filaments, rods, protective coatings, molded
13 and extruded shaped articles, and the like, by procedures
14 known in the art. These compositions are particularly use-
15 ful for producing materials requiring clarity, such as
16 blow-molded containers and the like, provided the melt is
17 quenched quickly.

18 This invention and its advantages will be better
19 understood from the following examples.

20 Example 1

21 Blends of various grades of EP elastomers and
22 homo polypropylene were Banbury mixed and underwater pel-
23 letized as shown in Table 1. The pellets were then fed
24 into a 1" 24/1 L/D 3 HP extruder and cast into film form
25 on a chilled roll. Optical properties of each film were
26 measured on a Gardner XL-211 Hazemeter. The characteris-
27 tics of each grade of EP elastomer are described in the
28 Table. The unmodified homo polypropylene was cast as a
29 control film. As shown by the data, optical properties
30 (haze and gloss) vary, depending on the particular grade
31 of elastomer used. Lowest haze and highest film gloss
32 were achieved with a 15 ML, 78% C₂ = EPDM. High haze was
33 generally observed with high Mooney elastomer. An ethyl-
34 ene content of 43 percent resulted in a tacky film sur-
35 face. Although the blends contain 60 percent elastomer,
36 their optics surprisingly rival the performance of unmodi-
37 fied homo polypropylene and the resulting films are very

1 flexible due to the high elastomer content.

2 Example 2

3 Blends of some of the same elastomers from Ex-
4 ample 1 were mixed with a random reactor copolymer poly-
5 propylene as shown in Table 2 and processed into film in
6 the same manner. Comparing the data between Tables 1 and
7 2 indicates slightly better optics for the reactor copoly-
8 mer vs. the homo polymer. Again, very good optics for
9 EP-4 grade of EPDM are noted relative to the other grades
10 tested, once again comparing favorably to the polyolefin
11 itself, but at significantly reduced stiffness.

12 Example 3

13 Pellets of EP-4 elastomer and a random reactor
14 copolymer (same as Example 2) were dry blended and fed to
15 an extruder and cast into film. Compositions evaluated as
16 shown in Table 3 demonstrate that good optical properties
17 were achievable with this grade of EPDM over a wide range
18 of blend compositions without the benefit of high shear in-
19 tensive mixing. The extruder used was a 1" Killion Model
20 KL-100 24/1 L/D.

21 Example 4

22 The blends shown in Table 4 were Banbury mixed
23 and underwater pelletized into approximately 1/8-inch pel-
24 lets. They were cast into film on a small laboratory ex-
25 truder, producing the excellent optical properties shown.
26 Bags were fabricated from the films using an impulse heat
27 sealer and filled with approximately 800 g. water. The
28 filled bags exhibited a soft feel and outstanding see-
29 through clarity. The bags were successfully autoclaved
30 without dimensional change at 250°F and retained a very
31 high level of see-through clarity after sterilization.

32 Example 5

33 The compositions of Example 4 were also success-
34 fully blow molded into a flexible container, filled with
35 water, and steam sterilized. The containers exhibited
36 good see-through clarity before and after autoclaving.

TABLE 1

Example 1

	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>	<u>E</u>	<u>F</u>
3 <u>Film</u>						
4 Homo PP (4.5 MFR)	40	40	40	40	40	100
5 EP-1	60	-	-	-	-	-
6 EP-2	-	60	-	-	-	-
7 EP-3	-	-	60	-	-	-
8 EP-4	-	-	-	60	-	-
9 EP-5	-	-	-	-	60	-
10 A.O.	0.3	0.3	0.3	0.3	0.3	-
11 Slip Agent	0.2	0.2	0.2	0.2	0.2	-
12 <u>Gauge, mils</u>	7.0	7.1	8.0	5.1	5.0	4.9
13 <u>Haze, %</u>	84	63	17	9	33	18
14 <u>Gloss, 45°, %</u>	3	6	-	36	23	-
15 <u>Comment:</u>	Non-Tacky	Non-Tacky	Tacky Surface	Non-Tacky	Non-Tacky	Non-Tacky
16	Tacky	Tacky	Tacky	Tacky	Tacky	Tacky
17	Surface	Surface	Surface	Surface	Surface	Surface

18 EP-1 : 50 ML (1 + 8) 127°C, 65% C₂=, Broad MWD EPDM
 19 EP-2 : 55 ML (1 + 8) 127°C, 70% C₂=, Narrow MWD EPDM
 20 EP-3 : 25 ML (1 + 8) 127°C, 43% C₂=, Narrow MWD EPDM
 21 EP-4 : 15 ML (1 + 8) 127°C, 78% C₂=, Broad MWD EPDM
 22 EP-5 : 50 ML (1 + 8) 127°C, 77% C₂=, Narrow MWD EPDM
 23 A.O. : Irganox 1076
 24 Slip Agent: Armoslip EXP

TABLE 2

Example 2

	<u>Film</u>	<u>G</u>	<u>H</u>	<u>I</u>	<u>J</u>
4	PPRC (4 MFR, 3% C ₂ =content)	40	40	40	100
5	EP-2	60	-	-	-
6	EP-4	-	60	-	-
7	EP-5	-	-	60	-
8	A.O.	0.3	0.3	0.3	-
9	Slip Agent	0.2	0.2	0.2	-
10	<u>Gauge, mils</u>	6.0	5.9	5.9	4.9
11	<u>Haze, %</u>	56	8	27	10
12	<u>1% Secant Modulus, psi-MD</u>	17,700	20,800	22,500	60,600

13 EP Description - See TABLE 1.

TABLE 3

Example 3

	<u>Film</u>	<u>K</u>	<u>L</u>	<u>M</u>	<u>N</u>	<u>O</u>
17	PPRC	80	70	60	50	40
18	EP-4	20	30	40	50	60
19	<u>Gauge, mils</u>	5.4	6.0	7.1	6.2	6.8
20	<u>Haze, %</u>	20	18	14	21	15
21	<u>Gloss, 45°, %</u>	28	33	38	34	46
22	<u>1% Secant Modulus</u>	42,300	39,400	33,300	24,000	22,700
23	<u>psi-MD</u>					

TABLE 4

Example 4

	<u>Film</u>	<u>P</u>	<u>Q</u>
27	PPRC	70	70
28	EP-4	30	-
29	EP-6	-	30
30	A.O.	0.3	0.3
31	Oxidized PE Wax	1.0	1.0
32	<u>Haze, %</u>	9	12

33 EP-4 : 15 ML (1 + 8) 127°C, 78% C₂=, Broad MWD EPDM

34 EP-6 : 23 ML (1 + 8) 127°C, 67% C₂=, Narrow MWD EPM

1 The advantages of this invention will be apparent
2 to the skilled in the art. Blends are provided that can be
3 processed in a broad variety of equipment to provide opti-
4 cally clear, impact resistant, tear resistant, durable
5 products. The products have excellent low temperature prop-
6 erties, exhibit easy heat sealing, and are quite flexible.
7 They may be sterilized by steam at 250°F without dimensional
8 change and without appreciable loss of optical clarity.

9 It will be understood that this invention is not
10 limited to the specific examples which are offered as par-
11 ticular embodiments, and that modifications can be made
12 without departing from the spirit thereof.

0120562

CLAIMS

1. A composition blend capable of being formed into optically clear products consisting essentially of:
 - (A) about 1 to 80 weight percent of an ethylene propylene elastomer having an ethylene content of at least 50 weight percent and a Mooney viscosity ML (1+8) at 127°C in the range of about 10 to 40; and
 - (B) about 20 to 99 weight percent of a polypropylene polymer selected from the group consisting of polypropylene and random polypropylene reactor copolymer.
2. A blend according to claim 1, wherein the elastomer has an ethylene content of from 60 to 80 weight percent.
3. A blend according to claim 1 or 2 wherein the elastomer has a Mooney viscosity of from 15 to 25.
4. A blend according to claim 1, 2 or 3, wherein the polypropylene is a random reactor copolymer.
5. A blend according to any of the preceding claims wherein the copolymer contains from 2 to 15 weight percent ethylene.
6. A blend according to any of the preceding claims, wherein the copolymer contains about 3 percent ethylene, and has a melt flow rate of 4.0 at 230°C.
7. A blend according to any of the preceding claims wherein the elastomer is ethylene, propylene, 5-ethylidene-2-norbornene.

8. A blend according to any of the preceding claims which additionally includes an incidental ingredient selected from the group consisting of process aids, lubricants, antiblocking aids, antioxidants, foaming agents, slip agents and mixtures thereof.
9. The use of a blend according to any of the preceding claims to form an optically clear product.
10. An optically clear product whenever produced from a blend according to any of the preceding claims.